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(54) PRODUCTION OF 2,6-DIMETHYLNAPHTHALENE AND OPERATION OF FILTER

(57)Abstract:

PROBLEM TO BE SOLVED: To stably in a long time obtain the subject high-purity compound capable of stably and continuously being filtered for a long period without causing clogging by cleaning a filter mediu after filtration of dimethylnaphthalene crystal.

SOLUTION: When 2,6-dimethylnaphthalene crystal is filtered, filter cake is released by a filter device for crystal slurry and a filter medium is cleaned with a solvent (preferably a solvent kept at a higher temperature than filter temperature). A rotary vacuum filter is preferably used as the filter medium. One or more kinds of solvents selected from an aliphatic saturated hydrocarbon and an alicyclic saturated hydrocarbon are preferably used as the solvent.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[The technical field to which invention belongs] this invention relates to 2, 2 [useful as a raw material of 6-naphthal dicarboxylic acid], the manufacture method of 6-dimethylnaphtalene especially 2, the filtration method of 6-dimethylnaphtalene crystal, and the operating method of a filter.
[0002]
[Description of the Prior Art] Especially 2 and 6-dimethylnaphtalene is useful as a raw material of 2 and 6-naphthale dicarboxylic acid, and 2 and 6-naphthalene dicarboxylic acid has an important use industrially as a raw material of th outstanding tensile strength and the highly efficient polyester used for manufacture of a polyethylenenaphthalate fibe a film, etc. which have thermal resistance. 2 and 6-dimethylnaphtalene (a dimethylnaphtalene is described as DMN below) oxidizes, serves as 2 and 6-naphthalene dicarboxylic acid, and depending on the case, dimethyl esterification carried out further and it serves as 2 and 6-naphthalene dicarboxylic-acid dimethyl.
[0003] 2 as a raw material of the highly efficient polyester used for manufacture of polyethylenenaphthalate fiber, a film, etc., 6-naphthalene dicarboxylic acid and 2, and 6-naphthalene dicarboxylic-acid dimethyl need to be the things a high grade, and 2 as the raw material and 6-DMN also need to be high grades. That is, although ten isomers exist in DMN with the position of two methyl groups, 2 as raw materials, such as 2 and 6-naphthalene dicarboxylic acid, and DMN need to be the things of the high grade which does not contain impurities, such as monomethyl naphthalene an trimethyl naphthalene, substantially further, either, excluding an isomer besides these substantially.
[0004] If the purity of 2 and 6-DMN is low, the impurity itself contained will oxidize, it will be esterified and purity, such as 2 and 6-naphthalene dicarboxylic acid, will be reduced. Moreover, it originates in the impurity in 2 and 6-DMN, and some impurities generated at oxidization and an esterification process are remarkably difficult to remove, and it becomes it is remarkable and difficult to obtain 2 of a high grade, 6-naphthalene dicarboxylic acid and 2, and 6 naphthalene dicarboxylic-acid dimethyl. Furthermore, existence of these impurities reduces notably the acid of not o the purity of an acid and ester but 2 and 6-DMN criteria, and the yield of ester. Therefore, in order to manufacture 2, naphthalene dicarboxylic acid and 2, and 6-naphthalene dicarboxylic-acid dimethyl on advantageous conditions industrially, it can be said that it is indispensable to obtain 2 of a high grade and 6-DMN.
[0005] 2 as an oxidization raw material and the purity of 6-DMN have the good higher one as much as possible. However, the load of refining becomes large in order to raise the purity of 2 and 6-DMN. Therefore, when refining a the influence on the load of refining as DMN, the results of oxidation reaction of DMN purity, and oxidization produ purity, further 2, and a 6-naphthalene dicarboxylic-acid dimethyl, 2 as an oxidization raw material and the purity of 6 DMN take into consideration the reaction results of methyl-ester-izing, the load of refining of a methyl ester, etc., an are decided. However, even if you take these into consideration synthetically, let purity of 2 and 6-DMN be at least 98.0% or more of high grade.
[0006] There is the method of carrying out isomerization separation, after methylating the method, the naphthalene, the methylnaphthalene separated from a tar fraction and a petroleum fraction as a manufacturing method of 2 and 6-DMN etc. These fractions and products need to separate 2 and 6-DMN from much isomer mixture including ten sort of most isomers. On the other hand, O-tolyl pentene -2 is cyclized to the method of obtaining O-tolyl pentene -2 from an ortho xylene and a butadiene by high yield to JP,49-134634,A, and JP,50-89353,A. The method of manufacturing and 5-dimethyl tetralin, and the method of carrying out the dehydrogenation of the 1 and 5-dimethyl tetralin to JP,48

76852,A, and manufacturing 1 and 5-DMN with high yield quantity selectivity are shown, to JP,50-129534,A, 1 and DMN are isomerized further, and 1, 5-, 1, 6, -, 2, and the method of obtaining the isomer mixture which consists of 6 DMN are mainly shown. Therefore, by combining these methods, 1, 5-, 1, 6, -, 2, and the isomer mixture that consis of 6-DMN can mainly be obtained from an ortho xylene and a butadiene, and there is also a method of separating 2 a 6-DMN from this isomer mixture.

[0007] It is necessary to separate and collect 2 and 6-DMN(s) from DMN isomer mixture by the 2 and 6-DMN manufacture method learned conventionally as mentioned above. It is very difficult to refine 2 and 6-DMN by distillation which the boiling point of dimethylnaphtalene 10 isomer is very close, and is usually well used for separation refining of an organic compound. After making 2, and 6-DMN and a complex form as 2 and the refining method of 6-DMN using the method by crystallization, the method by adsorption, and a certain kind of organic compound and separating this, the method of decomposing this complex etc. is proposed. Among these all directions methods, the method by crystallization is the simplest and it is suitable as the industrial refining method. Especially when mainly manufacturing 1, 5-, 1, 6, -, 2, and the isomer mixture that consists of 6-DMN and dissociating from an ortho xylene and a butadiene from now on, since there are few isomer kinds in a refining raw material, the method b crystallization is effective. When carrying out isomerization separation after methylating naphthalene, or when dissociating from a tar fraction and a petroleum fraction, it is necessary to separate 2 and 6-DMN from much isomer mixture, and the combination of an adsorption process and the crystallizing method is desirable in this case.

[0008] It is known that 2 and 6-DMN will form 1, 5-DMN, 2, 7-DMN, 2, and 3-DMN and an eutectic. Therefore, in order to deposit 2 and 6-DMN as a crystal by crystallization from isomer mixture, 2 in mixture and the quantitative ratio of 6-DMN and these isomers need to be larger than a eutectic composition ratio. That is, when 2 in mixture, 1 t 6-DMN, and the isomer mixture whose mole ratio of 1.4 or less, 2, and 3-DMN the mole ratio of 1.9 or less, 2, and 7 DMN is 1.1 or less for the mole ratio of 5-DMN are cooled, 2 and 6-DMN deposit first as a crystal.

[0009] As a way crystallization separates 2 and 6-DMN from DMN isomer mixture, by JP,50-22553,B, this mixture cooled and how to process the crystal which deposited with a methanol is shown. Moreover, JP,48-5767,A shows ho to wash or recrystallize this mixture using an aromatic hydrocarbon. However, a means to separate the crystal which deposited also in which these methods, and a mother liquor is not described in detail. Although it faces separating a crystal from the slurry formed of crystallization and the stability of the simple nature of a facility and operation and operation, high separation efficiency, etc. are required in order to carry out industrially, about the method of separati a 2 and 6-DMN crystal, what was indicated from such a viewpoint is not found from 2 which crystallized DMN isom mixture and was obtained, and a slurry including a 6-DMN crystal.

[0010]

[Problem(s) to be Solved by the Invention] This invention persons compounded 1 and 5-DMN by having used the or xylene and the butadiene as the raw material, isomerized this, and mainly got 1, 5-, 1, 6, -, 2, and the isomer mixture that consists of 6-DMN. And supplying continuously a jacket cooling formula crystallization tub with an agitator, af mixing this isomer mixture with a solvent, by passing cooling intermediation in a jacket, indirect cooling was carried out and the crystal of 2 and 6-DMN was deposited. However, when the crystal was continuously separated from the slurry which made the crystal of 2 and 6-DMN deposit, the filter cloth started blinding to the inside of a short time, a the problem that a filtration throughput will decline occurred. The purpose of this invention is a thing which 2 and 6-DMN are deposited by crystallization from DMN isomer mixture, and face detaching this by solid-liquid and manufacturing 2 of a high grade, and 6-DMN, is stabilized over a long period of time from the situation like a not les and can maintain a filtration throughput and for which an advantageous method is offered industrially.

[0011]

[Means for Solving the Problem] As a result of this invention persons' repeating examination wholeheartedly, the cau in which a filter cloth carries out blinding By the temperature of the liquid on a filter cloth falling, and being because crystal depositing within the stitch of a filter cloth, and washing the filter cloth after filtration with a solvent, when a solvent and a rinse evaporate, in case a crystallization mother liquor, and a solvent and a rinse are decompressed and attracted and are extracted It found out that it was stabilized and a filtration throughput could be maintained, and this invention was reached.

[0012] That is, this invention is the operating method of the filter characterized by washing a filter cloth using a hot solvent from filtration temperature in the manufacture method of 2 characterized by facing filtering the crystal of 2 a 6-dimethylnaphtalene and washing the filtering medium after filtration with a solvent, and 6-dimethylnaphtalene, an

the method of collecting crystals from a slurry. In addition, a rotary vacuum filter (it is henceforth described as RVF suitably used for filtration for obtaining a high grade 2 and 6-DMN from 2 and the crystallization slurry of 6-DMN. Moreover, as a solvent, a hydrocarbon especially an aliphatic saturated hydrocarbon, and an alicyclic saturated hydrocarbon are used suitably.

[0013]

[Embodiments of the Invention] The DMN isomer mixture with which crystallization is presented in this invention w not depend on the origin, if 1, 5-DMN, 2, 7-DMN, 2, and the composition ratio of 3-DMN to 2 and 6-DMN have 1.9 and 1.4 or 1.1 or less need, respectively and satisfy this composition ratio from a relation with the above-mentioned eutectic composition. In addition, although based also on the catalyst with which the DMN isomerization product compounded considering the ortho xylene and the butadiene as a raw material is used for isomerization, for 5 - 20%, and 6-DMN, 35 - 50%, 2, and 6-DMN is [1 and 5-DMN / the DMN isomer of 35 - 50% and others] usually 5% or less, the composition ratio of the above-mentioned [a such DMN isomerization product] is satisfied, and it can appl suitable for

[0014] Although the DMN isomer mixture with which are satisfied of the above-mentioned composition can be cool and 2 and 6-DMN can be obtained as a crystal, when DMN isomer mixture is cooled as it was, the 2 and 6-DMN crystal obtained is a scale-like, and its filterability is very bad. On the other hand, when a solvent is made to live together, it becomes a board-like crystal, and filterability is improved sharply. Therefore, it is more desirable to crystallize using a solvent, in order to obtain the crystal of a high grade by easy filtration and rinse operation. Use of solvent is indispensable in order to obtain the crystal which has 98% or more of especially purity. As a solvent, the filterable improvement effect has a greatly desirable hydrocarbon especially aliphatic saturated-hydrocarbon, and alicyclic saturated hydrocarbon. That is, as a solvent at the time of crystallization, the hydrocarbon especially aliphat saturated hydrocarbon, or alicyclic saturated hydrocarbon from the point of a crystallinity-like improvement, for example, a pentane, a hexane, a heptane, an octane, a nonane, Deccan, a methyl decane, a dimethyl decane, a cyclohexane, a methylcyclopentane, a methylcyclohexane, a cyclooctane, a decalin, a methyl decalin, a dimethyl decalin, etc. are desirable.

[0015] It is desirable to carry out filtration processing of the slurry obtained by crystallization continuously by RVF. RVF is equipment which performs solid liquid separation continuously by carrying out suction filtration, being immersed in a slurry and rotating a part of cylinder-like filtering medium (filter cloth). After a mother liquor is attrac by considering the interior of a cylinder as reduced pressure, the cake formed on the filter cloth by being immersed i slurry is washed by the suitable rinse, and are collected by exfoliating from a filter cloth further. Although a solvent a cake rinse are contained in the collected cake, 2 of a high grade and 6-DMN can be obtained by removing this solv and cake rinse by meanses, such as distillation. When RVF is used, solid liquid separation is not only made, but it ca perform each operation of the rinse of a cake, suction of a rinse, and cake exfoliation continuously, and there is an outstanding advantage that 2 of a high grade and 6-DMN can be manufactured by one step of operation of continuati Moreover, since rotational speed of RVF is low, it has few mechanical failures and also has the advantage that maintenance is easy.

[0016] In the solid liquid separation by RVF, exfoliation of the filter cloth to a slurry being immersed, suction of a mother liquor, the rinse of a cake, and a cake is performed one by one, and after the cake which was immersed in the slurry and extracted on the filter cloth has a crystallization mother liquor attracted, it is washed by the suitable rinse. is desirable to use for this rinse the same thing as what was used as a solvent on the occasion of crystallization. After that, a cake has a rinse attracted, further, exfoliates and are collected from a filter cloth. Thus, 2 of a high grade and 6 DMN can be obtained by removing a solvent and a rinse from the collected cake.

[0017] In this invention, after cake exfoliation before a filter cloth is again immersed in a slurry, a filter cloth is washed. By this washing, the 2 and 6-DMN crystal leading to blinding is dissolved without stopping operation of RV it is stabilized for a long period of time, and a filtration throughput can be maintained. This filter cloth washing operation may always be performed continuously, and may be intermittently performed according to the degree of th blinding of a filter cloth. The filter cloth penetrant remover used by this invention is a liquid under an operating condition, and it must have a crystallization mother liquor, a cake rinse, and compatibility at the same time it dissolv 2 and 6-DMN. Reactivity with further 2 and 6-DMN is low, and although there will be no limit especially if separati with DMN is easy, it is desirable to use the same thing as the solvent at the time of crystallization or a cake rinse from recovery of a crystallization mother liquor, a rinse, and a penetrant remover and the point of separation. An above-

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mentioned aliphatic saturated hydrocarbon and an above-mentioned alicyclic saturated hydrocarbon suitable at the ti of crystallization are equipped with all properties, like that reactivity with dissolving that they are the conditions with which a penetrant remover should be equipped, i.e., a liquid, 2, and 6-DMN, that there are a crystallization mother liquor, a cake rinse, and compatibility, 2, and 6-DMN is low, and separation with DMN is easy, and are suitably used as a penetrant remover.

[0018] Although there is especially no limit in the temperature of the filter cloth penetrant remover used by this invention, it is the purpose to dissolve the crystal which deposited by being cooled on a filter cloth, and filter cloth washing has [the temperature] the higher one more desirable than filtration temperature, i.e., slurry temperature. Sin a high cleaning effect is obtained by using a hot solvent for a filter cloth penetrant remover, the time of filter cloth washing can be shortened, and there is little amount of the penetrant remover used, and it comes to end. In addition, although the amount of penetrant removers around the unit time for obtaining a suitable cleaning effect is determined by the structure of filter cloth area or a filter, the amount of penetrant removers and washing time around this unit tim take a cleaning effect into consideration, and the optimal conditions are chosen.

[0019] In addition, although it is suitable when obtaining the crystal of 2 and 6-DMN, generally using the solvent which can dissolve a crystal as a filter cloth penetrant remover, in case a filter cloth is washed after filtering using filters, such as RVF, and exfoliating a cake like the above, or making the temperature higher than filtration temperatu i.e., slurry temperature, can be applied, when collecting not only this crystal system but crystals from a slurry especially. There is especially no limit in the crystallization tub for carrying out this invention, and the crystallization tub usually used for crystallization can be used.

[0020]

[Example] Next, an example explains this invention to a detail further. However, this invention is not limited to the following examples. In addition, it asked for the concentration of each component in the following examples with th gas chromatography.

[0021] Mixture of the composition shown in 1 manufactured from the example 1 ortho xylene and the butadiene and Table 1 which mixed the isomerization generation liquid and the normal heptane which isomerized and obtained 5-DMN, and was obtained was used as the crystallization raw material. This crystallization raw material was cooled at degrees C, and it considered as the slurry of 15 % of the weight of crystal concentration, and considered as the filtrat raw material. The crystal of 2 and 6-DMN was separated from the filtration raw material of 15 % of the weight of th 40-degree-C crystal concentration by the following methods, using RVF as a filter. The filter cloth is stretched on the front face by cylindrical [with a width of face / of 100mm /, and a diameter of 300mm], and, as for the filtration section of RVF, the filter cloth washing nozzle for washing a filter cloth after the rinse nozzle for washing a cake and cake exfoliation is attached in RVF.

[0022] The filtration raw material was supplied to RVF by the constant speed of 1000 kg/hr, and service conditions, such as a filter cloth drum rotational frequency, a filtration degree of vacuum, etc. of RVF, were adjusted so that the throughput of RVF might serve as 400 kg/hr. In the rinse, the cake was uniformly washed with the constant flow of 6 kg/hr using the 40-degree C normal heptane. The 40-degree C normal heptane was used with the constant flow of 30 kg/hr as a filter cloth penetrant remover after exfoliating a cake, and the filter cloth was always washed continuously was operated steadily in about 2 minutes after a filtration start, and operation was continued after that for 24 hours. T rotational frequency of RVF at the time of a stationary was 5rpm, and the filtration degree of vacuums were 500mmH (s). It did not generate, but the blinding of a filter cloth was able to be stabilized and was able to continue the filtratio processing by RVF in the meantime. The recovery of the crystal by filtration was about 92%. Although on stream an the separation crystal which exfoliates from RVF were sampled suitably and 2 under crystal and 6-DMN purity were investigated, the purity except the normal heptane was 99.0% or more.

[0023]

[Table 1]

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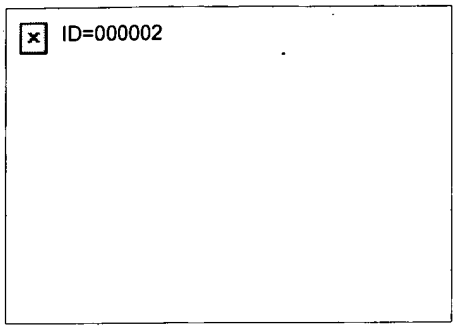

[0024] Although it was operated like the example 1 except not performing example of comparison 1 filter-cloth washing and filtration operation was started, however it might adjust the service condition of RVF when the filter clo started blinding and passed after the start up promptly for 30 minutes, the slurry filtration throughput stopped at 100 less kg/hr, and became uncontinuable [filtration operation].
[0025] It replaced with having performed example 2 filter-cloth washing continuously 30 kg/hr, always using a 40-degree C normal heptane, and using the 70-degree C normal heptane, except having carried out intermittently by the flow rate of 30 kg/hr for 2 minutes for every progress for 20 minutes, it was operated like the example 1 and filtratio operation was performed. It was operated steadily in about 2 minutes after a filtration start, and operation was continued after that for 24 hours. It did not generate, but the blinding of a filter cloth was able to be stabilized and wa able to continue the filtration processing by RVF in the meantime. The recovery of the crystal by filtration was abou 92%. Although on stream and the separation crystal which exfoliates from RVF were sampled suitably and 2 under crystal and 6-DMN purity were investigated, the purity except the normal heptane was 99.0% or more. making temperature of filter cloth washing into 40 to 70 degrees C to having performed filter cloth washing continuously in example 1 from the same effect being acquired by filter cloth washing for 2 minutes for every progress in the examp 2 for 20 minutes -- the amount of the filter cloth penetrant remover used -- about -- it turns out that it can consider as 1/10
[0026]
[Effect of the Invention] Without starting blinding by washing the crystal which washed the filtering medium after filtration and adhered to the filtering medium by the method of this invention so that clearly from the above example can be stabilized for a long period of time, and a filter (RVF) can be continued, it can operate, and a high grade 2 and 6-DMN crystal can be obtained to stability for a long time. Moreover, by washing a filtering medium at an elevated temperature by the method of this invention, the time of filter cloth washing is cut down and the amount of the penetrant remover used can be decreased. Therefore, since curtailment of you TERITI used for a filter by this invent and laborsaving of operation come to be measured and a high grade 2 and 6-DMN can be manufactured advantageou industrially, the industrial meaning of this invention is very large.

[Translation done.]